

Quantum entanglement of a tunneling spin with mechanical modes of a torsional resonator

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Abstract. - We solve Schrödinger equation describing a tunneling macrospin coupled to a torsional oscillator. Energy spectrum is studied for various quantum regimes. Magnetic susceptibility and noise spectrum are computed. We show that entanglement of the spin with mechanical modes of a subnanometer oscillator results in the decoherence of spin tunneling. For larger oscillators the presence of a tunneling spin can be detected through splitting of the mechanical mode at the resonance. Our results apply to experiments with magnetic molecules coupled to nanoresonators.

Introduction. – There has been enormous progress in measurements of individual nanomagnets [1], microcantilevers and microresonators [2–15]. Experiments have demonstrated that a mechanical torque induced by the rotation of the magnetic moment may be used for developing high-sensitivity magnetic probes and for actuation of micro-electromechanical devices. The underlying physics is a direct consequence of the conservation of the total angular momentum: spin plus orbital. While this side of the effect is clear, the mechanism by which the angular momentum of individual spins gets transferred to the rotational motion of a body as a whole has been less understood. In a macroscopic body it involves complex evolution of interacting spins and phonons towards thermal equilibrium. In that respect the case of a magnetic nano- or microresonator is simpler due to the great reduction of the number of mechanical degrees of freedom.

Recently, theoretical study of rotating magnetic nanosystems has been conducted within classical [16–19] and semiclassical [20,21] approaches. When spin is treated quantum-mechanically, further reduction of the number of degrees of freedom can be achieved in the low energy domain where only the lowest spin doublet that originates from the tunneling between spin-up and spin-down states is relevant. This would be the case of, e.g., a single-molecule magnet. Rigorous quantum-mechanical treatments have been recently suggested for the problem of a tunneling macrospin in a freely rotating body [22] and for

the problem of a tunneling macrospin coupled to the rotational modes of a nanoresonator [23] (see Fig. 1). These

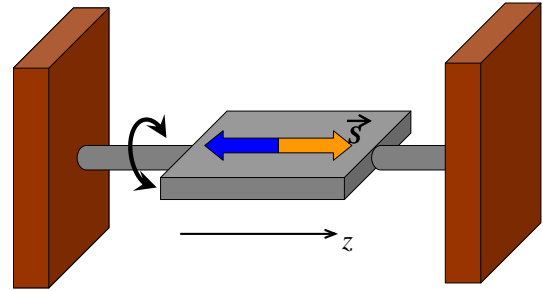


Fig. 1: System studied in the paper. Macrospin (e.g., a magnetic molecule) is attached to a torsional oscillator such that the magnetic anisotropy (quantization) axis is parallel to the axis of mechanical rotations.

two problems have one common feature: The spin tunneling becomes suppressed when the body containing the spin is too light. The physics behind this effect is quite clear [24]. Delocalization in the spin space that corresponds to tunneling of spin \mathbf{S} between spin-up and spin-down states reduces the energy by $\Delta/2$, where Δ is the splitting of the tunneling doublet. Since spin transitions are accompanied by the change of the angular momentum they generate

rotational motion of the body with the energy $\hbar^2 L^2/(2I)$, where I is the moment of inertia and L is the mechanical angular momentum that is generally of order S . At small I such rotations cost too much energy, so that the tunneling in the ground state should be frozen. This effect is conceptually similar to the decoherence and freezing of the tunneling of a particle in a double-well potential due to dissipation [25].

In this paper we solve Schrödinger equation for a macrospin coupled to a nanoresonator, Fig. 1. By considering various ranges of parameters of the nanoresonator, we reproduce previously obtained results and establish connection with the problem of a macrospin in a freely rotating body. Qualitatively different behavior is found for different ranges of parameters, that can be interpreted as a quantum phase transition. We show that the way to look for these effects is to study the electromagnetic response of the system depicted in Fig. 1. Our most important finding is that the coupling of a tunneling spin to a mechanical resonator destroys quantum coherence only for very small resonators – generally resonators consisting of just a few atoms. In resonators of greater size the coherence is preserved, and the presence of a tunneling spin can be detected by observing frequency splitting of mechanical oscillations.

The model. – Consider a model of a tunneling spin S , projected onto the lowest tunneling doublet, in a nanoresonator of torsional rigidity k that can rotate around the z -axis [21], see Fig. 1,

$$\hat{H} = \frac{\hbar^2 L_z^2}{2I_z} + \frac{I_z \omega_r^2 \varphi^2}{2} - \frac{W}{2} \sigma_z - \frac{\Delta}{2} (e^{-2iS\varphi} \sigma_+ + e^{2iS\varphi} \sigma_-). \quad (1)$$

Here $L_z = -i\partial_\varphi$ is the operator of the mechanical angular momentum, I_z is the moment of inertia of the resonator, $\omega_r = \sqrt{k/I_z}$ is the frequency of torsional vibrations, $W = 2Sg\mu_B B_z$ is the energy bias due to the longitudinal field B_z , Δ is the tunnel splitting of spin-up and spin-down states due to crystal field, and σ are Pauli matrices. As we shall see below, the behavior of such a system depends on two dimensionless parameters:

$$\alpha = \frac{2\hbar^2 S^2}{I_z \Delta}, \quad r = \frac{\omega_r}{\Delta}. \quad (2)$$

In the limit of a free particle, $r = 0$, the total angular momentum of the system with respect to the z axis is conserved: $J_z = S_z + L_z = \text{const}$. Tunneling of the spin changes S_z by $2S$, and this change is absorbed by the opposite change of L_z . Thus tunneling occurs between two quantum states having the same total angular momentum eigenvalue J . Computation of the eigenstates of the system reduces to the diagonalization of a 2×2 matrix. The resulting spectrum of the system has the ground state with $J = 0$ for [22]

$$\alpha \leq \alpha_1 = \left[1 - 1/(2S)^2\right]^{-1} \quad (3)$$

(heavy particle) that corresponds to the spin tunneling between up and down, with the change in the angular momentum absorbed by the rotation of the particle. However, for $\alpha > \alpha_1$ the ground state becomes degenerate and in the limit $\alpha \gg \alpha_1$ (light particle) it approaches $J = \pm S$, which means that the spin cannot tunnel.

In the case of a particle having a restoring force, that is the subject of this work, the total angular momentum of the spin and the mechanical oscillator is not conserved. Conservation of the angular momentum occurs in a larger closed system. Still, through the crystal field, tunneling of the spin generates mechanical torque acting on the torsional oscillator [21, 26]. This interaction can seriously reduce spin tunneling for both small and large r when the oscillator is light. In particular, for small r and large α (see below) the ground state is nondegenerate but the gap between the ground state and the first excited state becomes very small and the tunneling becomes effectively blocked. For this problem Kovalev et al. [23] introduced another dimensionless parameter,

$$\lambda = \sqrt{\frac{2\hbar S^2}{I_z \omega_r}} = \sqrt{\frac{\alpha}{r}}, \quad (4)$$

that is especially useful in the case of large r . One of their observations is that at $r \gg 1$ coupling of the spin to quantized rotational vibrations renormalizes the tunnel splitting according to

$$\Delta_{\text{eff}} = \Delta e^{-\lambda^2/2}. \quad (5)$$

To solve the quantum mechanical problem of a spin tunneling in a rotating body, it is convenient to use the basis that is a direct product of the two-state “up/down” basis for the spin and the harmonic oscillator basis for the body. Thus we write the system’s wave function $|\Psi\rangle$ in the form

$$|\Psi\rangle = \sum_{m=0}^{\infty} \sum_{\sigma=\pm 1} C_{m\sigma} |m\rangle |\sigma\rangle. \quad (6)$$

The coefficients $C_{m\sigma}$ satisfy the Schrödinger equation

$$i\hbar \frac{dC_{m\sigma}}{dt} = \sum_{n=0}^{\infty} \sum_{\sigma'=\pm 1} H_{m\sigma, n\sigma'} C_{n\sigma'}, \quad (7)$$

where

$$H_{m\sigma, n\sigma'} = E_{m\sigma} \delta_{mn} \delta_{\sigma\sigma'} - \frac{1}{2} \Delta_{\text{eff}} \times (\kappa_{mn} \delta_{\sigma, -1} \delta_{\sigma', 1} + \kappa_{mn}^* \delta_{\sigma, 1} \delta_{\sigma', -1}) \quad (8)$$

are matrix elements of the Hamiltonian, Eq. (1). Here

$$E_{m\sigma} = \hbar\omega_r(m + 1/2) - (1/2)W\sigma \quad (9)$$

are energies in the absence of tunneling and [23]

$$\kappa_{mn} = (i\lambda)^{m-n} \sqrt{\frac{n!}{m!}} L_n^{(m-n)}(\lambda^2) \quad (10)$$

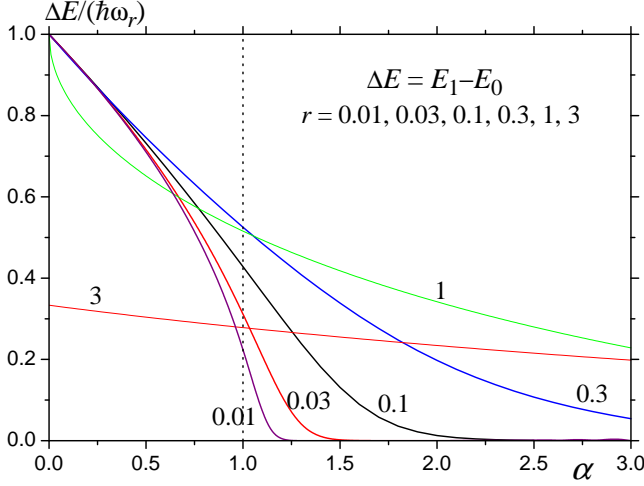


Fig. 2: Distance ΔE between the ground state and first excited state vs α for different r .

for $m \geq n$ and a similar expression with $m \Leftarrow n$ for $n \geq m$, where $L_n^{(m-n)}$ are a generalized Laguerre polynomials and λ is given by Eq. (4). In particular,

$$\kappa_{00} = 1, \quad \kappa_{10} = \kappa_{01} = i\lambda, \quad \kappa_{11} = 1 - \lambda^2. \quad (11)$$

For $r \gg 1$ only the ground state of the resonator is relevant in spin tunneling because energies of all other states are too high compared to Δ [23]. In this case one returns to a two-state model for the spin with the effective splitting (5). For small r , the spin couples to many oscillator states and one has to diagonalize a large matrix.

Energy spectrum and static susceptibility. – Setting $C_{m\sigma} \Rightarrow C_{m\sigma} e^{-i(E/\hbar)t}$ in Eq. (7) results in the stationary Schrödinger equation that can be diagonalized numerically to find energy eigenvalues E_μ . The results for the distance ΔE between the ground state and the first excited state vs α for different r and $W = 0$ are shown in Fig. 2. For $r \ll 1$ and $\alpha > 1$ the ground state becomes quasidegenerate with very small, although nonzero ΔE . This corresponds to the localization of the spin in either spin-up or spin-down state. On the other hand, ΔE does not exclusively characterize the spin but also contains information about the resonator. In particular, for $r < 1$ and $\alpha \rightarrow 0$ the spin and the resonator effectively decouple and $\Delta E \rightarrow \hbar\omega_r$, which is the mode of the resonator. On the other hand, for $r > 1$ and $\alpha \rightarrow 0$ one has $\Delta E \rightarrow \Delta$, which is the spin tunneling mode.

The spin susceptibility is

$$\chi = \frac{\partial \langle \sigma_z \rangle}{\partial W} \quad (12)$$

in the limit $W \rightarrow 0$. For a spin in a massive (non-rotating) body one has

$$\langle \sigma_z \rangle = \frac{W}{\sqrt{\Delta^2 + W^2}}, \quad (13)$$

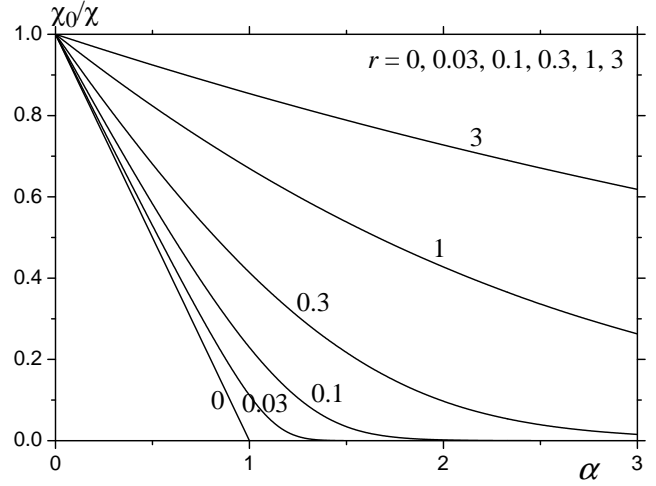


Fig. 3: Reduced inverse susceptibility vs α for different r .

thus the zero-field susceptibility is $\chi_0 = 1/\Delta$. For a spin in a rotating body, the effective splitting, Δ_{eff} , can be defined through $\chi = 1/\Delta_{\text{eff}}$, where $\chi = \partial \langle \sigma_z \rangle / \partial W$ follows from the exact numerical diagonalization of the Hamiltonian,

$$\langle \sigma_z \rangle = \sum_{m=0}^{\infty} \sum_{\sigma=\pm 1} \sigma |C_{0,m\sigma}|^2, \quad (14)$$

$C_{0,m\sigma}$ being the coefficients of the wave function corresponding to the ground state, $\mu = 0$. The dimensionless ratio $\chi_0/\chi = \Delta/\Delta_{\text{eff}}$ vs α for different r is shown in Fig. 3. For $r \ll 1$ and $\alpha > 1$ the zero-field susceptibility becomes very large because of quasidegeneracy of the “up” and “down” spin states. For $r \gg 1$ Eq. (5) is recovered.

Spin-rotation resonance. – The case $\sqrt{\Delta^2 + W^2} \approx \hbar\omega_r$ corresponds to the spin-rotation resonance that leads to a strong hybridization of spin and rotational states even in the case $\lambda \ll 1$. In the absence of the interaction between the spin and the resonator, $\lambda = 0$, the lowest four energy levels are

$$E = \left\{ \pm \frac{\sqrt{\Delta^2 + W^2}}{2}, \hbar\omega_r \pm \frac{\sqrt{\Delta^2 + W^2}}{2} \right\}, \quad (15)$$

where the zero-point energy of the resonator has been dropped. The hybridized levels are $\sqrt{\Delta^2 + W^2}/2$ and $\hbar\omega_r - \sqrt{\Delta^2 + W^2}/2$. The truncated low-energy Hamiltonian matrix has the form

$$\mathbb{H} = \begin{pmatrix} \frac{W}{2} & 0 & \frac{\Delta}{2} & \frac{\Delta}{2}i\lambda \\ 0 & \hbar\omega_r + \frac{W}{2} & \frac{\Delta}{2}i\lambda & \frac{\Delta}{2}(1-\lambda^2) \\ \frac{\Delta}{2} & -\frac{\Delta}{2}i\lambda & -\frac{W}{2} & 0 \\ -\frac{\Delta}{2}i\lambda & \frac{\Delta}{2}(1-\lambda^2) & 0 & \hbar\omega_r - \frac{W}{2} \end{pmatrix} \quad (16)$$

where Eq. (11) was used. We look for $E \approx \sqrt{\Delta^2 + W^2}/2 \approx \hbar\omega_r/2$. Then for $\lambda \ll 1$ the equation

$\det(\mathbb{H} - E\mathbb{I}) = 0$ simplifies to

$$\left(E - \frac{\sqrt{\Delta^2 + W^2}}{2}\right) \left(E - \hbar\omega_r + \frac{\sqrt{\Delta^2 + W^2}}{2}\right) = \frac{\lambda^2 \Delta^2}{4}. \quad (17)$$

At the resonance, $\hbar\omega_r = \sqrt{\Delta^2 + W^2}$, the frequencies of the transition between the ground state $E_0 = -\sqrt{\Delta^2 + W^2}/2$ and the closest excited states become

$$\omega_{\pm} = \frac{E - E_0}{\hbar} = \omega_r \left(1 \pm \frac{\lambda}{2} \frac{\Delta}{\sqrt{\Delta^2 + W^2}}\right). \quad (18)$$

This formula provides the splitting of the mechanical and spin modes at the resonance. For such a splitting to be observable, the quality factor of the mechanical resonator must exceed $(1 + W^2/\Delta^2)/\lambda$. Eq. (18) can also be obtained within semiclassical approximation [20].

Spin dynamics. — In the problem of a tunneling spin embedded in a non-rotating crystal, the parameter Δ has a clear physical meaning of the energy gap between the lowest tunneling doublet. When such a spin is prepared in, e.g., the spin-up state at $t = 0$, the probability to find it in the same state at an arbitrary moment of time t oscillates on t according to $\langle \sigma_z(t) \sigma_z(0) \rangle = \langle \sigma_z \rangle_t = \cos(\Delta t/\hbar)$. When the spin is coupled to a light oscillator and $r \gg 1$, one has to replace $\Delta \Rightarrow \Delta_{\text{eff}}$. At $r \ll 1$, coherent probability oscillations are destroyed at any non-zero α .

Spin dynamics is governed by the Schrödinger equation, Eq. (7), and the time dependence of $\langle \sigma_z \rangle$ is given by

$$\langle \sigma_z \rangle_t = \sum_{m=0}^{\infty} \sum_{\sigma=\pm 1} \sigma |C_{m\sigma}(t)|^2, \quad (19)$$

where $C_{m\sigma}(t)$ can be expanded over the eigenstates $C_{\mu;m\sigma}$ as

$$C_{m\sigma}(t) = \sum_{\mu} a_{\mu} \exp\left(-\frac{iE_{\mu}t}{\hbar}\right) C_{\mu;m\sigma}, \quad (20)$$

the coefficients a_{μ} being determined by the initial condition. If at $t = 0$ the spin was in the “up” state and the particle was in its ground state, one has $a_{\mu} = C_{\mu;01}^*$. Combining these formulas yields the time dependence

$$\langle \sigma_z \rangle_t = \sum_{\mu\mu'} A_{\mu\mu'} \exp\left(i\frac{E_{\mu} - E_{\mu'}}{\hbar}t\right), \quad (21)$$

where

$$A_{\mu\mu'} = a_{\mu}^* a_{\mu'} \sum_{m=0}^{\infty} \sum_{\sigma=\pm 1} C_{\mu;m\sigma}^* \sigma C_{\mu';m\sigma}. \quad (22)$$

Fourier spectrum of this time dependence, $2|A_{\mu\mu'}|$, gives the imaginary part of the susceptibility. Plotted vs $\hbar\omega_{\mu\mu'} = E_{\mu} - E_{\mu'}$, it gives an idea of the resonance absorption of the rf field by the spin. For $r \ll 1$ the Fourier spectrum consists, in general, of many lines. In the limit $\alpha \rightarrow 0$ the spin and the torsional oscillator decouple; in this case only one line of height 1 remains. For $\alpha \ll 1$

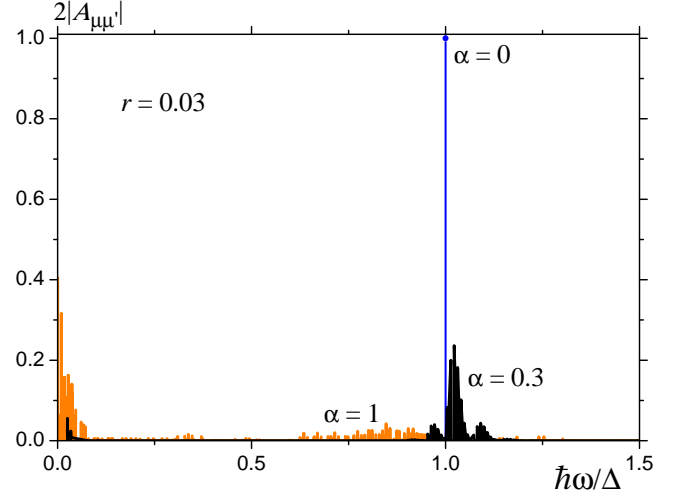


Fig. 4: Fourier spectrum of $\langle \sigma_z \rangle_t$ for $r = 0.03$ and $\alpha = 0, 0.3$, and 1.

there is a narrow group of lines with a spread that gives rise to spin decoherence due to interaction of the spin with the oscillator. At $\alpha > 1$ decoherence becomes very strong and the low-frequency part of the Fourier spectrum corresponding to mechanical oscillations becomes large. These results are shown in Fig. 4 for $r = 0.03$.

At $r \gg 1$, there is only one line of height 1 at $\hbar\omega = \Delta_{\text{eff}}$ with Δ_{eff} given by Eq. (5). This is natural because in this limit the problem is described by an effective two-state model. For $r = 1$ and small α there is a doublet of lines around $\hbar\omega = \Delta$ because of the resonance interaction between the spin and the resonator.

Discussion. — We have studied energy spectrum, susceptibility, and decoherence in a system consisting of macrospin rigidly coupled to a torsional mechanical resonator. Our general conclusion is that the coupling does not influence spin tunneling when the resonator is sufficiently large and heavy. However, when one approaches the atomic size the magneto-mechanical coupling may lead to strong decoherence of the spin states. To put these statements in perspective, let us consider a magnetic molecule of spin 10 embedded in a torsional resonator in the shape of a paddle of dimensions $20 \times 20 \times 10 \text{ nm}^3$. As in Ref. [23] we shall assume that the paddle is attached to the walls by two carbon nanotubes of torsional rigidity [27] $k = 10^{-18} \text{ N}\cdot\text{m}$. The moment of inertia of such a system is dominated by the paddle, $I_z \sim 10^{-36} \text{ kg}\cdot\text{m}^2$, so that $\omega_r = \sqrt{k/I_z} \sim 10^9 \text{ s}^{-1}$. The parameter λ is then of order 10^{-2} . For $\Delta/\hbar \ll 10^9 \text{ s}^{-1}$, coherent spin oscillations at frequency Δ/\hbar will not be affected by the coupling to the paddle. They will be more likely decohered by the coupling to nuclear spins or other environmental degrees of freedom in the same manner as for a spin embedded in

a macroscopic solid. For $\Delta/\hbar > 10^9 \text{s}^{-1}$ the parameter r will be small. However, α will be very small compared to one, and, thus, in accordance with Fig. 4, no decoherence of spin oscillations due to coupling with the mechanical oscillations of the paddle will occur in this case either. The same will be true even if instead of the paddle one considers, e.g., a Mn_{12} molecule attached to a carbon nanotube [28]. The relevant moment of inertia is now that of the molecules itself, which for a nanometer size molecule is of order $10^{-42} \text{kg}\cdot\text{m}^2$. The corresponding ω_r is of order 10^{12}s^{-1} and $\lambda \sim 0.1$. The two regimes are now $r \gg 1$ for $\Delta/\hbar \ll 10^{12} \text{s}^{-1}$ and $r \ll 1$, $\alpha \ll 10^{-2}$ for $\Delta/\hbar \gg 10^{12} \text{s}^{-1}$. In both limits the mechanical oscillations should have little effect on coherent spin oscillations with frequency Δ/\hbar .

The above estimates show that the effects on tunnel splitting and spin decoherence arising from spin-rotation coupling should not be much of a concern in nanomechanical setups with large magnetic molecules that have been discussed in literature. To have a significant effect on the tunnel splitting one should arrive to $\lambda > 1$. This requires much smaller moments of inertia, that is, molecules much smaller than Mn_{12} . Consider, e.g., a small magnetic molecule of spin 10 with the moment of inertia that is one hundred times smaller than that of the Mn_{12} molecule. We shall also assume that it is coupled to the walls with the torsional rigidity one hundred times smaller than the coupling through a carbon nanotube. In this case one still gets $\omega_r \sim 10^{12} \text{s}^{-1}$ but $\lambda > 1$. Now the regime with $r \sim 1$ is achieved at $\Delta/\hbar \sim 10^{12} \text{s}^{-1}$, which corresponds to $\alpha \sim 1$. In this case one should expect significant decoherence of spin oscillations. The bottom line is that decoherence due to a resonator may occur in subnanometer systems but it should not be pronounced above the nanometer size. This is easy to understand if one notices that $I_z \omega_r$ in the expression $\lambda = S \sqrt{2\hbar/I_z \omega_r}$ is the measure of the “macroscopicity” of the resonator. Consequently, $I_z \omega \sim \hbar$ needed to achieve large λ generally requires a system of the atomic size. For larger resonators, interaction between spin and mechanical degrees of freedom reveals itself only near the resonance. It results in a very interesting quantum phenomenon that can be observed in experiment: Splitting of the mechanical mode of the resonator containing a tunneling spin. Indeed, in our example with a paddle having $\lambda \sim 10^{-2}$ the splitting of the mechanical mode at the resonance can be quite significant. For $\Delta < \hbar \omega_r$ the resonance will be achieved at $W/\hbar \sim \omega_r \sim 10^9 \text{s}^{-1}$, which for $S = 10$ corresponds to the magnetic field of order 10G. For, e.g., $\Delta/\hbar \sim 10^8 \text{s}^{-1}$, according to Eq. (18), this will provide the splitting in the MHz range that would be possible to observe if the quality factor of the resonator exceeds one thousand.

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